

Correlated Alteration Effects in CM Carbonaceous Chondrites

Lauren B. Browning^{1,3}, Harry Y. McSween, Jr.¹, and Michael E. Zolensky²

¹ Department of Geological Sciences, University of Tennessee, Knoxville,
TN 37996-1410, USA

² Earth Science and Solar System Exploration Division, NASA Johnson
Space Center, Houston, TX 77058, USA

³ *Present address:* Hawaii Institute of Geophysics and Planetology, School
of Ocean and Earth Science and Technology, University of Hawaii at
Manoa, 2525 Correa Road, Honolulu, HI 96822, USA

GCA MS 10628

Abstract - Three parameters are proposed to determine the relative extent of alteration in CM chondrites. The mineralogic alteration index monitors the relative progress of coupled substitutions in the progressive alteration of cronstedtite to Mg-serpentine, and increases with increasing alteration. To calculate values of this index, an algorithm has been developed to estimate the average matrix phyllosilicate composition in individual CM chondrites. The second parameter is the volume percent of isolated matrix silicates, which decreases with progressive alteration due to mineral hydration. Finally, the volume percent of chondrule alteration monitors the extent of chondrule phyllosilicate production, and increases as alteration proceeds. These parameters define the first CM alteration scale that relies on multiple indicators of progressive alteration. The following relative order of increasing alteration is established by this model: Murchison \leq Bells $<$ Pollen \leq Murray $<$ Mighei $<$ Nogoya $<$ Cold Bokkeveld. Bulk $\delta^{18}\text{O}$ values generally increase with progressive alteration, providing additional support for this sequence. The relative degree of aqueous processing Cochabamba and Boriskino experienced is less precisely constrained, although both fall near the middle of this sequence. A comparison between the mineralogic alteration index and literature values of the whole-rock chemistry of CM chondrites reveals several correlations. For example, a positive, nearly linear correlation between bulk H content and progressive CM alteration suggests an approximately constant production rate of new phyllosilicates relative to the mineralogical transition from cronstedtite to Mg-serpentine. Furthermore, the abundance of trapped planetary ^{36}Ar decreases systematically in progressively altered CM chondrites, suggesting the wholesale destruction of primary noble gas carrier phase(s) by aqueous reactions. Multiple correlations between diverse alteration parameters indicate that different CM chondrites probably experienced the same kinds of processes and conditions during in situ parent body alteration.

INTRODUCTION

CM carbonaceous chondrites are widely believed to sample incompletely serpentinized fragments of asteroidal bodies containing some of the most primitive materials known to man. As such, these meteorites offer a rare glimpse at the effects of nebular, accretionary, and asteroidal processing on early solar system materials. Unfortunately, the perplexing combination of petrologic features and chemical properties that these meteorites exhibit greatly hinders attempts to answer many fundamental questions regarding their origin.

Over the years, there has been considerable disagreement over the location where CM materials experienced alteration. Early workers advocated the direct condensation of hydrated CM materials out of cooling nebular gases (GROSSMAN and LARIMER, 1974; BARSHAY and LEWIS, 1976), but theoretical models later showed that silicate hydration is likely to have been kinetically inhibited in the nebula (PRINN and FEGLEY, 1987). Renewed attention was then given to the parent body alteration model, which had developed earlier from extensive petrologic observations (MCSWEEN, 1979; BUNCH and CHANG, 1980; BARBER, 1981; TOMEOKA and BUSECK, 1985). This model asserts that the alteration of CM materials occurred in situ on the CM parent body when accreted ices melted and reacted with co-accreted anhydrous materials (GRIMM and MCSWEEN, 1989). Additional asteroidal processes, such as regolith gardening events, further contributed to the petrologic complexity of CM materials. This model has recently been challenged, however, by the preaccretionary alteration model (METZLER et al., 1992), which advocates the completion of aqueous alteration in the solar nebula, the subnebula of Jupiter, or, on special precursor CM planetismals. Following an extensive petrological study of CM chondrites, these authors concluded that CM rim textures formed in the nebula when partially altered chondrules and other coarse-grained core components passed through one or more nebular dust clouds containing mostly fine-grained hydrated

phases. The CM parent body then accreted from the agglomeration of these rimmed components, and experienced little or no subsequent alteration. Clearly, these 2 opposing models submit fundamentally different scenarios of early solar system history, and a resolution is needed.

If CM alteration occurred in situ, many intrequeing avenues are opened toward an improved understanding of early solar system processes and materials. Because individual CM chondrites have experienced varying degrees of incomplete alteration (MCSWEEN, 1979; BUNCH and CHANG, 1980; TOMEOKA and BUSECK, 1985), for example, the chronology of secondary reactions that effected them might be deduced by evaluating reaction textures and mineral associations that were arrested in various stages of development. Knowledge of this reaction sequence is essential for an improved understanding of the evolving conditions and materials on C-type asteroids, the most logical source of CM chondrites (KERRIDGE and BUNCH, 1979).. In addition, the alteration reactions effecting CM materials ought to provide a useful analog for other types of solar system materials that experienced interactions between melted nebular ices and anhydrous minerals. For example, the nuclei of short-period comets, ice-bearing satellites, and many interplanetary dust particles may be thoroughly modified by aqueous alteration processes that are analogous to those which affected carbonaceous chondrite parent bodies (MCSWEEN and WEISSMAN, 1989). Furthermore, several meteorites appear to have textures and mineralogies that are transitional between the incompletely altered CM and thoroughly altered CI chondritic classes (IKEDA and PRINZ, 1993; ZOLENSKY and BROWNING, 1994) , suggesting a relationship that may best be elucidated by extrapolation from CM, rather than CI, samples. Moreover, some CV and unequilibrated ordinary chondrites (HUTCHISON et al., 1987; TOMEOKA and BUSECK, 1990) contain small amounts of phyllosilicates, implying that these largely anhydrous meteorite classes may have experienced alteration events that were similar to, but less extensive than, those effecting CM chondrites. Consequently, it would be advantageous to determine whether

the diverse material properties of CM chondrites can be explained by predictable alteration trends that indicate well-behaved reaction progress on the CM parent body.

We emphasize the need to establish a reliable scaling procedure for determining the relative degree of alteration that different CM materials exhibit. Without a clearly defined alteration scale, the task of unraveling corroborating alteration trends from the motley collection of material properties that different CM chondrites possess can only proceed haphazardly. Unfortunately, however, the identification of reliable progressive alteration parameters is not a trivial exercise. Since the number of asteroidal source(s) for CM chondrites is unknown, for example, it is possible that some petrologic features of individual CM samples simply reflect differences inherent on multiple CM parent bodies. Moreover, uncertainties involving the nature of the original CM materials and the effects of asteroidal accretion and resurfacing (HOUSEN et al., 1979) generally limit the applicability of terrestrial serpentinites as CM analogs. Finally, the fine grain size of CM alteration phases leads to unavoidable analytical problems, while the wide range of textures and mineral associations observed within a single CM thin section readily lends itself to varied, and often self-contradictory, interpretations regarding their origin.

Because many uncertainties complicate the fundamental distinction between progressively altered CM chondritic samples, a dependable scaling procedure must employ multiple parameters that allow the identification of self-consistent trends. These alteration parameters should gauge the influence that different degrees of aqueous processing have had on the diverse textural, mineralogical, isotopic, and bulk chemical properties of individual CM chondrites (CLAYTON and MAYEDA, 1984; VAN SCHMUS and WOOD, 1967; ZOLENSKY et al., 1993). As such, an integrated set of scaling parameters ought to provide a reliable framework for the addition of new alteration parameters.

In this paper, we introduce the first relative alteration scale for CM chondrites that relies on multiple correlated parameters. Quantitative data involving modal proportions

and average phyllosilicate compositions from 9 CM falls define 3 progressive alteration indicators that are then used to identify 3 additional alteration trends based on bulk chemical and isotopic compositions. The simultaneous employment of the proposed progressive alteration indicators produces a high-resolution scale, while the identified correlations themselves suggest additional constraints on the location and conditions of CM alteration.

SAMPLES AND ANALYTICAL TECHNIQUES

We obtained samples of 9 CM chondritic falls that are reported to vary widely in their extent of aqueous alteration (MCSWEEN, 1979). Table 1 lists the meteorite names, abbreviations, thin section numbers, and sources of the CM chondrites in this study. To minimize the effects of overlapping phases in the third dimension, petrographic thin sections were prepared with thicknesses roughly between 20 and 25 μm , when possible. Modal data was collected from these thin sections in reflected and transmitted light using a standard petrographic microscope equipped with a point counting stage. The total number of points counted for each sample was at least 1000.

We obtained chemical analyses of CM matrix material using a Cameca CAMEBAX electron microprobe at the NASA Johnson Space Center, operated at 15 kV with a beam current of ~ 30 nA. Standard data-reduction software was used to correct X-ray intensities for deadtime, background, and ZAF matrix effects. Natural minerals were used as standards. These wavelength dispersive analyses are considered to be accurate to within $\sim \pm 0.5\%$. Qualitative analyses of matrix material were also obtained on this microprobe, which is equipped with an energy-dispersive spectroscopy (EDS) detector system and scanning electron imaging capabilities.

PREVIOUS RESEARCH: MINERAL ALTERATION IN CM MATRICES

CM chondrites contain a wide variety of phases (ZOLENSKY and MCSWEEN, 1988) that are arranged into a smorgasbord of different textural components (BUNCH and CHANG, 1980): While even the most ostensible of textural components may be more difficult to distinguish from one another than previously thought (METZLER et al., 1992), one of these, the CM matrix, has long been discussed in the literature (MCSWEEN, 1977; BARBER, 1981; ZOLENSKY et al., 1993). MCSWEEN (1977) operationally defined the CM matrix, as "dark, aphanitic material free of coarse silicate grains, magnetite, sulfide, or metal optically discernable at 400 x in reflected light using a petrographic microscope". Matrix material is widely considered to be the volumetrically dominant (MCSWEEN, 1979; GREENWOOD et al., 1994; ZOLENSKY et al., 1993) and most extensively altered (MCSWEEN, 1977; BUSECK and XIN, 1993) textural component of CM chondrites. Thus, resolving progressive alteration trends in CM chondrites requires an awareness of the diverse mineral assemblages in CM matrices and the variety of processes that are capable of producing them. Much progress has been made in this area, and we summarize here only those results which have contributed directly to the development of our proposed alteration scale.

CM matrices contain a complex disequilibrium assemblage of minerals. Serpentine group minerals, saponite, tochilinite, tochilinite-serpentine intergrowths, olivine, pyroxene, various Fe-monosulfides, Fe-Mg oxides, carbonates, phosphates, Fe-Ni metal and amorphous material may all co-exist within a single CM matrix (ZOLENSKY et al., 1993). Of these, phyllosilicates may be the most reliable indicators of progressive CM alteration because the structures and compositions of these secondary minerals strongly reflect those of the precursor silicate grains, the interacting fluids, and the environmental conditions that accompany aqueous alteration (MOODY, 1976). Thus, a rigorous characterization of matrix phyllosilicates is essential for the identification of mineralogical trends with advancing CM alteration.

Since most matrix phyllosilicates in CM chondrites have grain sizes on the order of a micron or less, individual grains are generally studied by high-resolution techniques. Previous TEM analyses reveal a wide range of phyllosilicate compositions, structures, and morphologies (BARBER, 1981; TOMEOKA and BUSECK, 1985; ZOLENSKY et al., 1993). Individual grains are commonly intergrown with sulfides, tochilinite, carbonates, or other phyllosilicates, indicating an origin by complex, in situ replacement processes (BUSECK and XIN, 1993). ZOLENSKY and MCSWEEN (1988) reviewed the various types of phyllosilicates that have been reported in CM chondrites and argued that these are best described as belonging to the serpentine, berthierine, chlorite, and smectite groups. Of these, the serpentine group minerals are the most volumetrically significant (MCSWEEN, 1987). Table 2 provides a list of the formulae of the mineral endmembers within the serpentine group.

High-resolution mineralogical characterizations are often complemented by alternative techniques that may be better-suited for the identification of progressive alteration trends on a larger scale. For example, MCSWEEN (1979) proposed that the apparent content of phyllosilicate-rich matrix material in different CM chondrites should gauge the relative extent of phyllosilicate production and, therefore, the relative degree of aqueous alteration that individual CM chondrites experienced. He compared defocussed beam analyses of optically homogeneous matrix with the modal matrix contents of individual CM chondrites to determine if progressive alteration is accompanied by compositional changes. The matrix Fe/Si ratios from different CM chondrites were found to roughly decrease with increasing modal percentages of matrix and, hence, the relative degree of aqueous alteration. Similarly, BUNCH and CHANG (1980) noted that the Mg/Fe ratios of matrix phyllosilicates probably increase during advancing alteration, and BARBERS' (1981) microtextural analyses of CM matrices support the late formation of Mg-rich alteration phases.

It has long been established that terrestrial serpentinization typically involves simultaneous phyllosilicate formation and recrystallization (WICKS and WHITTAKER, 1977). Since the composition of every phyllosilicate in a system must reflect the state of the surrounding environment during its formation, the bulk Fe/Si and Mg/Fe trends reported earlier by MCSWEEN (1979) and BUNCH and CHANG (1980) demonstrate the large-scale evolution of matrix phyllosilicate compositions during progressive CM alteration. Microprobe analyses of CM matrices indicate that phyllosilicate compositions usually fall within a region that joins endmember cronstedtite and serpentine on a SiO₂-MgO-FeO ternary (MCSWEEN 1987; BROWNING et al., 1991). However, the range of matrix phyllosilicate compositions for different CM chondrites varies significantly within this field (ZOLENSKY et al., 1993). Although Fe³⁺ cannot be directly analyzed by microprobe techniques, the presence of variable amounts of Fe³⁺-rich phyllosilicates is supported by the lattice spacings of individual phyllosilicate grains (BARBER, 1981) and confirmed by Mössbauer analyses (BURNS and FISHER, 1993). Moreover, spectroscopic analyses support the presence of Fe³⁺ phyllosilicates on the surfaces of some C-type asteroids (VILAS et al., 1993). HRTEM observations (TOMEOKA and BUSECK, 1985) and mass balance calculations of bulk matrix compositions (MCSWEEN, 1987) provide a plausible explanation for the range of phyllosilicate compositions in CM matrices; progressive alteration is accompanied by a continuous mineralogic trend from Fe³⁺-rich cronstedtite to Mg-rich serpentine.

PROGRESSIVE ALTERATION SCALE

Our scaling procedure for determining the relative alteration of CM chondrites assumes a mineralogic transition from cronstedtite to serpentine with advancing alteration, and capitalizes on observed variations in the range of matrix phyllosilicate compositions and the degree that anhydrous silicates have been altered. In this section,

we introduce 3 new alteration parameters for determining the relative degree of aqueous alteration that individual meteorites have experienced. The first of these, the mineralogic alteration index, will subsequently be compared to other alteration parameters.

Mineralogic Alteration Index

Since endmember phyllosilicate compositions are relatively uncommon in CM matrices (ZOLENSKY et al., 1993), minerals with intermediate compositions best reflect the degree of CM alteration. The proposed mineralogic alteration index gauges the number of incremental steps that have been taken along the alteration pathway from cronstedtite to serpentine based on an approximation of the average matrix phyllosilicate composition for a given CM chondrite. To determine the mineralogic alteration index, we employ a descriptive definition for CM matrix, rather than an interpretive one (e.g. METZLER et al., 1992). CM matrix is defined here as the sum of fine-grained phases that forms an aphanitic groundmass between coarse-grained components, lithic clasts, tochilinite-rich clumps and rim textures. This modified version of the matrix definition given by MCSWEEN (1977) excludes specific CM components that are commonly embedded in or adjacent to the CM matrix, for additional clarity.

The continuous mineralogical transition from cronstedtite to serpentine requires that recrystallized and newly-formed phyllosilicates systematically discriminate between various cations as CM alteration proceeds. This is most clearly evidenced by the large differences in these endmember phyllosilicate compositions (Table 2). Ferric Fe replaces one tetrahedral Si and also occupies an octahedral position in the endmember cronstedtite structure. In addition to Fe^{3+} , meteoritic cronstedtite grains incorporate up to 5 wt.% Al (BARBER, 1981), which probably occupies a portion of the structural sites that are traditionally filled by Fe^{3+} . At the other extreme of the CM mineralogic trend, the idealized serpentine structure contains a full ration of tetrahedral Si and incorporates no

trivalent cations. In order to maintain an approximate mineralogic charge balance throughout the transition from endmember cronstedtite to serpentine compositions, this progressive alteration trend must have been accomplished by a sequence of coupled substitutions that systematically introduced tetrahedral Si and divalent octahedral cations into matrix phyllosilicates at the expense of Fe^{3+} and, to a lesser degree, Al.

We propose that progressive CM alteration is best monitored by evaluating the progress of Si and Fe^{3+} substitutions that necessarily attend the transition from endmember cronstedtite to serpentine. To track the redistribution of the relevant cations, we first employ a generalized stoichiometric phyllosilicate formula, $(\text{Fe}^{2+}, \text{Mg}, \text{Mn}, \text{Ca} \dots)_{3-x}(\text{Al}, \text{Fe}^{3+})_x(\text{Si}_{2-x}(\text{Al}, \text{Fe}^{3+})_x\text{O}_5(\text{OH})_4)$, to characterize the compositions of individual matrix phyllosilicates. This mineral formula accommodates the entire range of serpentine compositions observed in CM matrices (Table 2). The proposed mineralogic alteration index, $2 - (\text{Fe}^{3+}/(2-\text{Si}))$, is intended to isolate the relevant exchange information stored in the stoichiometric phyllosilicate formula. The enclosed ratio, $\text{Fe}^{3+}/(2-\text{Si})$, gauges the extent to which the coupled substitution of $2(\text{Fe}^{3+}, \text{Al}) = \text{Si} + (\text{Mg}, \text{Fe}^{2+} \dots)$ was completed in the average matrix phyllosilicate of an individual CM chondrite. The denominator of this ratio indicates the number of trivalent atoms that replace Si in the idealized serpentine structure, while the numerator is an approximation of the total number of Fe^{3+} atoms occupying tetrahedral and octahedral sites. An endmember cronstedtite composition would be represented, then, by a $\text{Fe}^{3+}/(2-\text{Si})$ ratio of 2. In contrast, an endmember serpentine composition always yields a $\text{Fe}^{3+}/(2-\text{Si})$ ratio of 0 because it contains 2 Si atoms and no Fe^{3+} . Hence, the value of this ratio decreases as alteration proceeds. By subtracting the ratio from 2, the mineralogic alteration index for a CM sample, $2 - (\text{Fe}^{3+}/(2-\text{Si}))$, is forced to *increase* as alteration proceeds. This was done for clarity, and does not effect the sensitivity of the enclosed ratio. An index value of 0 denotes pure cronstedtite and a minimum degree of alteration, while a value of 2 indicates pure serpentine and extensive alteration.

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A detailed account of the algorithm used to calculate values of the mineralogic alteration index is given in the Appendix, but we briefly summarize the procedure here. Phyllosilicate formulae were estimated from electron microprobe analyses of matrix material in each CM chondrite by characterizing all matrix phyllosilicates according to the generalized phyllosilicate stoichiometry described above. Since the microprobe beam is generally larger than the size of a single serpentine grain, we impose the requirement that each matrix analysis used in this study must reflect the composition of a mineral assemblage comprised exclusively of phyllosilicates. Our procedure involves the use of multiple screening devices which exclude those analyses that contain unwanted carbonate or oxides inclusions. More than half of our original matrix analyses were eliminated from further consideration because these or additional criteria were not met. Despite these efforts, small quantities of ubiquitously occurring S-bearing inclusions had to be subtracted from each analysis. For simplicity, we assumed that all S was incorporated into pyrrhotite inclusions. Although pyrrhotite probably is the most common S-bearing phase in CM matrices (ZOLENSKY, 1992), all analyses with ≥ 2 wt. % S were then discarded to limit the associated errors. Although the unavoidable presence of S in CM matrix analyses inevitably biases the inferred Fe content of the investigated phyllosilicates, microprobe-based investigations involving serpentine compositions in CM matrices rarely address this problem. After selecting our most reliable microprobe data, the number of Fe^{3+} atoms in each matrix analysis was estimated from measured concentrations of SiO_2 , Al_2O_3 , and FeO , by allowing matrix phyllosilicates to fulfill stoichiometric requirements. Individual values of $\text{Fe}^{3+}/(2\text{-Si})$ were calculated for each analysis. These were then averaged and subtracted from 2 to obtain the mineralogic alteration index for a given meteorite.

Table 3 gives the average serpentine formulae and mineralogic alteration indices for the CM chondrites in this study. Standard deviations for each cation demonstrate the

inherent compositional variability of individual serpentine grains within each meteorite, and are not an indication of analytical errors.

Relative Proportion of Isolated Anhydrous Silicates in CM Matrices

Olivine and pyroxene grains with diameters ranging from ~5 - 100 μm are commonly imbedded within the submicron-sized alteration assemblages that comprise CM matrices. These isolated silicate grains may have been derived from impact-fragmented chondrules (MCSWEEN, 1977; RICHARDSON and MCSWEEN, 1978; JONES, 1992). Since fluid infiltration should be efficient in an unconsolidated asteroidal regolith, anhydrous silicate grains in this type of environment would be highly susceptible to fluid-rock reactions. Indeed, many isolated anhydrous silicate grains in CM matrices show clear signs of having been partially altered by aqueous reactions. The surface of the olivine in Figure 1, for example, has been etched and pitted by alteration reactions that produced phyllosilicates and minor sulfides. Although the alteration features in Figure 1 are commonly observed in CM matrices, however, the isolated matrix grains in a single CM thinsection may range from unaltered to fully altered. The juxtaposition of unaltered and altered matrix silicates poses an interesting interpretive problem that is discussed in a section below.

We suggest the possibility that the modal proportion of anhydrous matrix silicates in different CM matrices provides another, independent indication of the relative degree of CM alteration. To test this parameter, the modal proportion of anhydrous matrix silicate grains in 9 different CM chondritic falls was obtained by point counting. Results of this analysis are plotted against the mineralogic alteration index in Figure 2a. Error bars in Figure 2a illustrate estimated 1σ counting errors, and were derived from the results of VAN DER PLAS and TOBI (1965). The method that these authors propose for estimating counting errors from modal data is valid under the condition that the step

interval between modal points is larger than the "largest grain fraction". In contrast, we used the smallest possible step interval between points to maximize the coverage of each thin section. Had we used their method, we would have counted approximately 4 times less points in a given area. VAN DER PLAS and TOBI (1965) take the reliability of modal results as $\sigma = [p(100-P)/n]^{1/2}$, where p = the real content of a mineral in percent by volume, P = the vol. % of a mineral obtained by modal counting, and n = the total number of points counted. Since number of points that we have counted, n' , will always be 4 times that of VAN DER PLAS and TOBI (1965) for a given area, we approximate the ratio of their error to our own error as $(n'/n)^{1/2}$.

Relative Percentage of Chondrule Alteration

There is no reason to suppose that every olivine and pyroxene grain in a chemically active asteroidal regolith would have been equally susceptible to aqueous reactions. Both the low surface/volume ratio and the coarse-grained, interlocking igneous textures within chondrules, for example, would have effectively discouraged the infiltration of alteration fluids into chondrule interiors. It is no surprise, then, that the ratio of unaltered to altered silicates within CM chondrules is apparently larger than that of the surrounding fine-grained matrix or rim components. However, if differences in the extent of chondrule alteration relative to matrix alteration are systematic, then a correlation should exist between the relative degree of alteration that the chondrules and isolated matrix silicates experienced.

We propose that the percentage of chondrule alteration phases should reflect the progressive consumption of anhydrous chondrule silicates by aqueous reactions. The relative percentage of chondrule alteration phases in different CM chondrites is expected, therefore, to increase with increasing alteration. Values for this chondrule alteration parameter were obtained by point counting the chondrule phases from 9 different CM

chondrites. The extent of chondrule alteration was then calculated as 100X the ratio of the number of modal points of phyllosilicate-bearing assemblages in chondrules divided by the sum of all chondrule points. The volume percentage of chondrule alteration is plotted against the mineralogic alteration index in Figure 2b. Error bars show the results of propagating counting errors across the ratio given above.

Variability and Uncertainty in Model Parameters

KERRIDGE (1985) argued that the heterogeneous nature of highly brecciated CM chondritic meteorites precludes the determination of accurate error values on analytical data. We applaud the integrity of this conclusion and submit, then, that the analytical errors presented in Figures 2 and 3 should be viewed as estimations of individual sources of error, rather than a precise account of the sum of all influencing sources of error. Although this situation is not ideal, the validity of our results is not diminished. Systematic errors in the 3 proposed alteration parameters should be vanishingly small due to the collection of all data at a single laboratory by one investigator who adhered to a common procedure for each sample. Hence, we are able to emphasize the significance of relative, rather than absolute, parameter values for individual CM chondrites. We outline here some intangible sources of error in the 3 proposed alteration parameters and describe our efforts to minimize their effects.

It is not possible to quantify errors associated with the calculation of mineralogic alteration indices. Most notably, the abundance of Fe^{3+} is an integral component of the index, but no significance can be ascribed to the absolute values of Fe^{3+} calculated in this work. However, since we use the same algorithm to calculate all Fe^{3+} values, systematic errors are minimized. Furthermore, we submit that the soundness of the logic used in devising this algorithm is demonstrated by the capacity of the mineralogic index to produce predictable alteration trends. Hence, we conclude that the index provides a

sensitive gauge of the *relative* progress that individual CM chondrites have made along the progressive alteration pathway from cronstedtite to Mg-serpentine.

Biases resulting from the heterogeneity of CM samples may be realized most clearly in modal values that were obtained from a limited number of thin sections. However, every effort has been made to maximize the reliability of our modal data. For example, we have deliberately selected modal-based alteration parameters in which the relevant phases are easily distinguished by standard petrographic techniques. Although fine-grained and mineralogically similar matrix and rim components must be distinguished to determine the proportion of isolated matrix grains for one parameter, so few anhydrous silicates are visible in rim material under an optical microscope that occasional judgement errors would have negligible impact on the results. Hence, both modal parameters essentially require only that a distinction be made between fine-grained alteration assemblages and coarse-grained anhydrous silicates. In addition, we evaluated the volume percent of CM chondrules that is occupied by alteration phases to limit the influence of sampling biases that may otherwise result from different thin sections of the same meteorite having variable proportions of chondrules. Very small counting distances were employed to ensure full coverage of each thin section and to maximize the total number of collected points, which improved the statistical significance of our results.

BULK CHEMICAL SIGNATURES OF PROGRESSIVE CM ALTERATION

In this section, we specifically address the possibility that the interacting alteration fluids themselves might have generated observable changes in the bulk chemical and isotopic signatures of CM chondrites that reflect the influence, evolution, and abundance of the reacting fluids. Three types of whole rock data are discussed here which may complement the mineralogical trends suggested above.

Bulk H Content

Since hydrous phyllosilicates volumetrically dominate the mineralogy of CM chondritic meteorites (MCSWEEN, 1987), it follows that a positive correlation might also exist between the advancement of CM alteration and the bulk H abundance (MCSWEEN, 1979), which reflects the H_2O or OH^- content of the rock. However, it should not be tacitly assumed that the total volume of interacting fluids will be represented by values of bulk rock H content. It is conceivable, for example, that advancing fluid-rock reactions may have been accompanied by phyllosilicate recrystallization and the redistribution of dissolved components, rather than by the generation of new phyllosilicates. In addition, BROWNING et al. (1995) explain how the armouring of anhydrous silicates by dense sulfide rind deposits might inhibit the continued alteration of precursor grains in CM chondrites. An positive correlation between the bulk H content and the alteration index number is expected to occur if both parameters are sensitive to the production of phyllosilicates.

All bulk H contents in this study were taken from the literature (VAN SCHMUS and HAYES, 1974; KERRIDGE, 1985). To counter some effects of sampling heterogeneity, we utilized Si-normalized values of bulk H. In some cases, Si-normalized values were not reported directly in the literature and had to be derived from the bulk H ($\mu\text{g/g}$) values and chemical compositions of the relevant CM chondrites. A numerical approximation of errors in the bulk H abundance of CM chondrites cannot be justified (KERRIDGE, 1985), although potential sources of error include analytical limitations, intra-meteorite heterogeneity and, perhaps, systematic inter-laboratory biases. Figure 3a illustrates the relationship between the mineralogic index and bulk H content in CM chondrites.

Abundance of Trapped Noble Gases

Trapped noble gases are present in most types of meteorites, but the abundance of these gases is highest in carbonaceous chondrites (SCHULTZ and KRUSE, 1989). The trapped gas content is subdivided into solar gas patterns, which are attributed to solar wind implantation, and planetary noble gas patterns, which are characterized by an enhancement of the heavy inert gases relative to the lighter gases (SWINDLE, 1988). Planetary noble gases are believed to have largely been incorporated into the meteorite when it formed (MAZOR et al., 1970). However, both the origin of planetary gas fractionation patterns and the cause of variations in the trapped gas abundances in different meteorites remain uncertain. We explore here the possibility that progressive CM alteration may have been accompanied by the systematic loss of trapped planetary gases.

Aqueous reactions resulted in the wholesale dissolution of many types of primary mineralogic and organic phases in CM chondrites (ZOLENSKY and MCSWEEN, 1988). This suggests that progressive alteration may have been accompanied by the systematic loss of planetary gases as carrier phases were consumed by aqueous reactions. With the complete destruction of a ^{So}carrier phase, all retained gases would have been released into the CM fluids. Hence, these gases are likely to have maintained their original fractionation patterns. Because noble gases are highly insoluble in H₂O (STUTE et al., 1992), it is reasonable to suppose that the alteration fluids would have provided an efficient medium to transport these inert components to the surface of the CM parent body. This is consistent with suggestions that the CM alteration system was open to the loss of at least a few components, such as Cl, CH₄, H₂O, and Na (BUNCH and CHANG, 1980; WOLF et al., 1980; ZOLENSKY et al., 1989). Since progressive alteration is characterized by the continued destruction of primary phases, we propose that the absolute abundance of planetary gases in different CM chondrites may reflect the relative degree of aqueous processing that individual meteorites experienced.

We take the abundance of ^{36}Ar as an approximation of the overall trapped planetary gas content in a CM chondrite. Given the relative abundance pattern of planetary gases (MAZOR et al., 1970), comparisons between the degree of CM alteration and the abundance of ^{84}Kr , ^{132}Xe , or ^{36}Ar are expected to produce roughly similar trends. The mean concentration of ^{36}Ar was obtained for most CM chondrites in this study from a compilation of noble gas data (SCHULTZ and KRUSE, 1989). In most cases, literature values of whole-rock ^{36}Ar were used. However, only analyses of light and dark material separates are available for Nogoya. Based on a photograph of the Nogoya sample in HEYMANN and MAZOR (1966), we visually estimated that the light material constituted 25 vol % of the analyzed sample, and weighed the data accordingly. However, BLACK's (1972) data for the light and dark Nogoya separates were averaged because no indication was given of their relative proportions. To avoid extensive biases resulting from averaging a small number of ^{36}Ar analyses from compositionally heterogeneous samples, any individual data point that did not fall within 2 standard deviations from the average of the remainder of values for that meteorite was excluded from this investigation. Since the only two whole rock values of ^{36}Ar for Murchison differed significantly, we estimated an additional bulk ^{36}Ar value for this meteorite by using our modal data to weigh values of ^{36}Ar in anhydrous silicates and matrix from Murchison separates. Figure 3b is a plot of the mineralogic alteration index versus the mean ^{36}Ar abundance (SCHULTZ and KRUSE, 1989). Uncertainties in the mean ^{36}Ar content of each meteorite are graphically depicted.

Oxygen Isotopic Ratios

The mineral fractions from CM chondrites display a greater variability in oxygen isotopic composition than any other type of meteorite (CLAYTON, 1993). Chondrule and CAI separates both plot on a ^{16}O -mixing line that probably reflects high temperature

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exchange with nebular gas. In contrast, CM matrices exhibit a significant enrichment of ^{17}O and ^{18}O that may have been incurred during low temperature fluid-rock exchanges, which typically occur with large mass-dependent fractionation factors. CM Chondrules contain mostly anhydrous silicates, and may be the source of similar silicate grains that are embedded in the CM matrix (MCSWEEN, 1977; RICHARDSON and MCSWEEN, 1978; JONES, 1992). In contrast, CM matrices are volumetrically dominated by hydrous phyllosilicates and are mineralogically similar to CM rim materials (ZOLENSKY et al., 1993). Hence, there is a clear distinction between the isotopic compositions of anhydrous and hydrous silicates which dominate the mineralogy of CM chondrites. Because the proportion of hydrated phases must increase with progressive CM alteration, the relative extent to which individual bulk CM samples are enriched in $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ may provide additional insight into the relative degree of alteration experienced by these samples. To test this hypothesis, we plot the available $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ data for bulk CM samples in this study, and compare their relative positions on the resultant mixing line, of slope 0.6 ± 0.04 . All oxygen isotopic data for the CM chondrites in this study were obtained from R. N. Clayton. Results are shown in Figure 3c. Error bars are smaller than the data points.

RESULTS AND DISCUSSION

We note that although the proposed alteration parameters monitor fundamental changes that are likely to accompany simple serpentinization processes, predictable trends for the aqueous alteration of CM chondrites are far from self-evident. CM materials are likely to have experienced a multiplicity of processes that could potentially thwart the reliability or interpretation of the progressive alteration parameters. For simplicity, the interpretation of our results given below assumes in situ parent body alteration. However, in a later section, we evaluate all aspects of our model to determine

if these are consistent with the most plausible of alternative explanations, preaccretionary alteration (METZLER et al., 1992).

Correlated Alteration Trends

Figure 2a implies a causal relationship between the relative degree of in situ alteration that an individual meteorite experienced and the modal content of isolated matrix silicates. This well-defined correlation strongly suggests that each CM chondrite in this analysis must have originally contained a similar proportion of isolated matrix silicates. Hence, the original abundance of isolated silicate grains in each sample was probably at least as high as that of Murchison, but was later diminished to varying degrees depending on the extent of aqueous alteration experienced by the individual CM chondrite. Likewise, the correlation in Figure 2a argues against the addition of significantly different amounts of anhydrous silicate grains to individual CM chondrites after the onset of aqueous alteration. Throughout the alteration process, each of the investigated samples probably maintained a roughly equivalent production rate of matrix silicate grains due to the disruption of chondrules during regolith gardening processes.

The percentage of chondrules that is replaced by phyllosilicate-rich assemblages increases during the mineralogical transition from cronstedtite to serpentine (Figure 2b). The chondrule alteration parameter reveals nothing about the absolute abundance of chondrules in different CM samples. However, in a manner similar to that used for matrix silicates, we argue that the systematic behavior of the data in Figure 2b would probably not have occurred if variable proportions of unaltered chondrules had been randomly added to different CM chondrites after aqueous alteration had already begun. Thus, the flux rate of unaltered chondrules to the CM regolith(s) was probably nearly identical for all samples.

Phyllosilicates were continually produced throughout the course of progressive CM alteration. The trends in Figure 2 demonstrate a volumetric increase in phyllosilicates as anhydrous silicates are systematically consumed by progressive aqueous reactions. Moreover, the nearly linear increase in the bulk H content of individual CM chondrites in Figure 3a suggests a roughly constant production rate of new hydrous phases relative to the mineralogical transition from cronstedtite to serpentine. A collective evaluation of Figures 2 and 3a indicates, then, that the evolving compositions of both newly-formed and recrystallized phyllosilicates were very sensitive to the abundance and oxidation state of dissolved Fe in the fluids throughout the course of CM alteration.

The positive correlation between the median bulk ^{36}Ar content and the mineralogic alteration index of individual CM chondrites (Figure 3b) is best explained by significant degassing caused by progressive alteration reactions. It seems less likely, for example, that this trend is the result of accreting decreasing proportions of noble gas carrier phases into asteroidal parent bodies that would fortuitously experience increasing degrees of aqueous alteration at some later time. Although different analyses of the planetary gas content in a single CM chondrite may vary widely (SCHULTZ and KRUSE, 1989), Figure 3b illustrates that the effect of sample heterogeneity was not significant enough to destroy the evidence for systematic variations between individual CM chondrites. As above, this trend suggests that the original planetary gas content of different CM chondrites was remarkably similar.

The correlation in Figure 3b indicates that the original planetary gas carrier phase(s) for CM chondrites was relatively susceptible to aqueous alteration and might have even been a major phase. Although only a small fraction of the original planetary gas content may currently reside in/on secondary carrier phases produced by fluid-rock reactions, at least some of these may be incorporated either into the structures or onto the surfaces of recrystallized or newly-formed phyllosilicates. Phyllosilicates are relatively

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efficient at adsorbing noble gases onto their surfaces (REYNOLDS et al., 1978) and incorporating a variety of mineral inclusions (ZOLENSKY and MCSWEEN, 1988) and organic products into their structures (CRONIN and CHANG, 1993).

Given the well-behaved correlations between the mineralogical alteration index and all other parameters discussed above, we conclude that each of the proposed indices is sensitive to the extent of CM alteration. Moreover, the reliability of the mineralogic alteration index provides additional support for previous assertions (BUNCH and CHANG, 1980; MCSWEEN, 1987; TOMEOKA and BUSECK, 1985) that progressive CM alteration is accompanied by a mineralogical transition between meteoritic cronstedtite and serpentine.

On the Relative Order of Increasing CM Alteration

Robust alteration parameters must not only monitor the progress of mineralogic, textural, and chemical trends in CM chondrites, but must also produce consistent assignments of the degree of alteration for specific meteorites. We begin this section by reviewing the relevant results of earlier investigations, and conclude with a discussion of the degree to which specific CM chondrites have been altered, as judged by this study.

Many conflicting suggestions have been offered regarding the relative order of increasing CM alteration. Based on the relative modal content of matrix materials in individual CM chondrites, for example, MCSWEEN (1979) suggested the following order of increasing aqueous alteration for the CM chondritic samples in this study: MY (58.8%) < MI (60.7 %) < MC (63.6%) < CB (74.2%) < BO (75.8%) < PO (80%) < BL (81.7%) < NG (85.4%). Following independent petrographic analyses of Bells (BL), BREARLEY (1994) agreed that this meteorite is extensively altered, but DAVIS and OLSEN (1984) did not. In addition, GREENWOOD et al. (1994) obtained a modal matrix content for Cold Bokkeveld of 82.7 vol. %, suggesting that this meteorite is significantly more altered than

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suggested by MCSWEEN (1979). TOMEOKA and BUSECK (1985) supported MCSWEEN's (1979) conclusion that Mighei, Murchison, and Murray are relatively unaltered compared to other CM chondrites, and provided additional petrographic evidence in support of a previous suggestion (BUNCH and CHANG, 1980) that CM matrix phyllosilicates become progressively enriched in Mg and depleted in Fe as alteration proceeded. ZOLENSKY et al. (1993) tested the proposals of both MCSWEEN (1979) and TOMEOKA and BUSECK (1985) by comparing average $\text{FeO}/(\text{FeO}+\text{MgO})$ ratios for the matrix phyllosilicates in several different CM chondrites with independently-determined modal values of CM matrices in these same samples. The $\text{FeO}/(\text{FeO}+\text{MgO})$ ratios were found to increase in the order: NG < MC < CB < MI < MY, and no correlation was identified between values of $\text{FeO}/(\text{FeO}+\text{MgO})$ and the modal matrix content of individual CM chondrites. An alternative scaling parameter, the relative abundance of Cl, was offered by BUNCH and CHANG (1980), who asserted the following order of increasing alteration: MC (1400 +/- 200 ppm Cl) < My (1000 +/- 150 ppm Cl) < CB (700 +/- 100 ppm Cl) < NG (400 +/- 50 ppm Cl). In addition, BURGESS et al. (1991) submitted that the oxidized/reduced S ratios of individual CM chondrites should increase with the extent of progressive aqueous alteration, which results in the trend MI (1.3) < MY (1.4) < MC (2.3) < NG (3.3). It was, in part, the lack of agreement in these studies that prompted us to re-evaluate the types of progressive alteration parameters that might best gauge advancing CM alteration.

Each of our proposed alteration parameters yields largely self-consistent predictions regarding the relative order of increasing CM alteration. Given the variety of factors that may have influenced the progressive alteration signatures of individual CM chondrites, however, it is not surprising that some uncertainties persist. Therefore, we note firstly that the correlations described in this work allow the unambiguous assignment of the investigated samples to the categories of minimal (MC and BL), intermediate (PO, MY, CC, BO, MI), and extensive (NG and CB) aqueous alteration. In addition, each of the progressive alteration trends demonstrate the following relative order of increasing

alteration for 7 of the 9 CM chondrites in this study: $MC \leq BL < PO \leq MY < MI < NG < CB$. The degree of aqueous processing that Cochabamba (CC) and Boriskino (BO) experienced, relative to the other intermediately-altered CM chondrites, is not clearly indicated by our data. However, the extent of Boriskino's alteration appears, on average, to be roughly equivalent to that of Mighei (MI), while Cochabamba (CC) is always more altered than Murray (MY).

Although segments of some earlier relative alteration scales for CM chondrites are reproduced by our progressive alteration parameters, the order of increasing CM alteration that is predicted by our model is generally inconsistent with previous results. Perhaps most notably, our model indicates a very low relative degree of alteration for Bells, but a recent TEM analysis has shown that the matrix mineralogy of this meteorite appears to have closer affinities to the more extensively altered CI chondrites (BREARLEY, 1995). Because magnetite is abundant and cronstedtite is absent in Bells (BREARLEY, 1995), magnetite inclusions may have contributed additional Fe to our matrix analysis that was not accounted for in our calculation of the average matrix phyllosilicate composition for this meteorite. Consequently, the low mineralogic alteration index value that we obtained for Bells implies the presence of abundant Fe-rich cronstedtite. Regardless, we note that the other model parameters, as well as the whole rock alteration indicators, establish Bells as a one of the least altered CM samples in this study. Because the sequence of alteration reactions that effected Bells clearly followed a different pathway than it did in other CM chondrites, geochemical modeling is needed to elucidate the special conditions under which the unusual Bells mineralogy formed.

Finally, it is instructive to compare the relative order of increasing CM alteration that is predicted by our model to the order of increasing $\delta^{18}\text{O}$ in bulk CM samples for which data are available. Figure 3c shows that Bells (BL) and Murchison (MC) have the smallest bulk $\delta^{18}\text{O}$ values, indicating the lowest degree of mineral hydration, while the most extensively altered sample, Cold Bokkeveld (CB), has the largest value of $\delta^{18}\text{O}$.

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Isotopic compositions thus provide additional support for the relative degree of alteration that is predicted for these meteorites by our model parameters. Unfortunately, no additional insight is gained for the other CM samples in Figure 3c because their $\delta^{18}\text{O}$ contents are poorly resolved. Similar discrepancies in the order of CM alteration that is predicted by a few of our own alteration parameters, as well as the collective parameters of previous researchers, caution that a reliable alteration scale for CM chondrites cannot be based on a single parameter. Thus, it is the multiply correlated parameters in our model that establish the general reliability of our results.

On the Location of CM Alteration

In an earlier section, the correlations between model alteration parameters were interpreted in the context of the parent body alteration model, which emphasizes the in situ alteration of anhydrous materials on the CM parent body or bodies. We now ask whether the existence of either the progressive alteration parameters themselves or the various correlations between these parameters might just as easily be explained by an alternative model involving the preaccretionary alteration of CM materials (METZLER et al., 1992). The alteration history of petrologically diverse clasts in polymict CM breccias are similarly evaluated based on our model results.

The individual alteration parameters in this work provide no new insight into the location of CM alteration. Irrespective of whether alteration was preaccretionary or in situ, the samples in hand contain CM chondritic materials that are aqueously altered to varying degree. Not surprisingly, then, there exists rational parameters that clarify the distinction between more altered and less altered materials. Of the various alteration parameters proposed in this work, only the mineralogic alteration index is strictly empirical. Having no known underlying rational of its own, then, the sensitivity of this

parameter to progressive CM alteration must be established by comparison with the other parameters.

In contrast, the multiple parameter correlations described in this paper are difficult to reconcile with the preaccretionary alteration model (METZLER et al., 1992). This model advocates the somewhat random association of altered and unaltered phases in CM chondrites due to physical mixing processes that occurred during nebular rim-forming events, the accretion of the CM parent body, and subsequent impact gardening processes. It would be unreasonably fortuitous for these multi-stage mixing events to produce different CM materials that seem to reflect variable progress along a well-behaved alteration pathway. Yet, our model parameters yield corroborating predictions for the relative extent of alteration experienced by individual CM samples by monitoring such diverse alteration properties as the volumetric proportions and compositions of hydrated phases. Thus, the correlations described in this paper reveal a measure of consistency in the alteration properties of each CM sample which strongly suggests that adjacent CM materials responded to the same alteration event. As such, our results provide compelling support for the parent body alteration model.

A separate, but related, topic of interest involves the location where the materials in CM clasts experienced aqueous alteration. Polymict CM breccias contain clasts that are petrologically distinct from the host material and these often exhibit variable degrees of aqueous alteration. Our analysis of the polymict CM breccias Cold Bokkeveld and Nogoya (ZOLENSKY and BROWNING, 1994; METZLER, 1995) reveal average matrix phyllosilicate compositions that correlate well with diverse whole rock alteration parameters. Moreover, our alteration parameters consistently indicate that both of these meteorites are extensively altered relative to other CM samples. These whole rock alteration parameters must largely reflect the weighted average of individual clast properties. Because self-consistent correlations exist for these polymict breccias, we strongly suggest that a) the matrix material in these meteorites contains representative

proportions of the various observed clasts, and b) most of the individual CM clasts themselves possess self-consistent alteration properties that are best attributed to parent body alteration reactions.

While the correlated alteration parameters described herein provide strong support for the parent body alteration model, we note that in situ alteration processes alone cannot explain the full range of characteristics that CM chondrites exhibit. Instead, CM materials preserve clear evidence of having been effected by a variety of nebular, accretionary, and secondary asteroidal processes. Because it is likely that individual CM components were effected to varying degrees by each of these processes in ways that are poorly understood, additional consideration ought to be given to the origin of specific CM features that are consistent with the preaccretionary alteration model. A comprehensive evaluation of these features is beyond the scope of this paper and will be treated separately in a forthcoming paper.

SUMMARY

We proposed 3 new indices for gauging the relative extent of alteration that individual CM chondrites experienced. The mineralogic alteration index gauges the relative extent to which the progressive alteration trend from cronstedtite to Mg-rich serpentine (BUNCH and CHANG, 1980; TOMEOKA and BUSECK, 1985; MCSWEEN, 1987) has been completed in individual CM chondrites. The proposed mineralogic alteration index monitors the incremental progress of coupled substitutions along this alteration pathway, and is based on an approximation of the "average" matrix phyllosilicate composition in individual CM chondrites. The second proposed parameter, the volume percent of isolated matrix silicate grains, evaluates the relative extent to which the anhydrous silicates in different CM chondrites were consumed by aqueous reactions. The proportion of altered phases within chondrules defines the third alteration parameter,

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which is sensitive to the volumetric production of chondrule phyllosilicates as hydration reactions proceed. Hence, we propose the first alteration scale for CM chondrites that relies on multiple parameters. Correlations between the proposed parameters illustrate a systematic decrease in the volume percent of anhydrous matrix silicate grains and a commensurate increase in the extent of chondrule alteration during the mineralogic transition from cronstedtite to Mg-serpentine.

The potential of advancing fluid-rock reactions to influence whole-rock chemical and isotopic signatures suggests the possibility of additional alteration trends in CM chondrites. Using bulk-rock data obtained from the literature, we tested the following hypotheses: 1) the abundance of bulk H reflects the extent of phyllosilicate production and, hence, the relative degree of aqueous alteration that individual CM chondrites experienced, and 2) the destruction of primary noble gas carrier phases by aqueous reactions resulted in systematic degassing of unfractionated trapped planetary gases during progressive CM alteration. A Comparison between the mineralogic alteration index and these bulk-rock signatures produced the predicted correlations. A nearly linear increase in bulk H content with progressive CM alteration implies an approximately constant production rate of phyllosilicates relative to the mineralogical transition from cronstedtite to Mg-serpentine. In addition, the loss of planetary trapped gases in progressively altered CM samples suggests that the original carrier phase for these gases was highly susceptible to aqueous reactions. More generally, the predictable behavior of all progressive alteration indicators indicates that individual CM chondrites may have been subjected to the same type of asteroidal processes and conditions as alteration proceeded. Finally, the multiply correlated alteration parameters provide strong evidence for the in situ aqueous alteration of CM materials on an asteroidal parent body.

An evaluation of all proposed progressive alteration indicators clearly demonstrates the following relative order of increasing CM alteration: Murchison \leq Bells $<$ Pollen \leq Murray $<$ Mighei $<$ Nogoya $<$ Cold Bokkeveld. Boriskino and Cochabamba

are probably best described as having experienced intermediate degrees of aqueous processing. Although less well constrained, the oxygen isotopic composition of bulk CM chondrites provides additional support for these predictions. Several discrepancies between the relative order of alteration predicted by the alteration parameters demonstrate that it is not possible to produce a progressive alteration scale based on a single parameter.

Acknowledgements - Funding for this work was provided by NASA Grant ??? to H. Y. McSween, NASA Grant 152-11-40-23 to M. Zolensky, and NASA Grant NAGW-3281 to K. Keil. We also thank the NASA Graduate Student Researchers Program for funds supplied to L. Browning through Grant ??/ to H. Y. McSween. Constructive reviews by J. Kerridge, I. Wright, and K. Metzler significantly improved this manuscript, and are greatly appreciated. We also thank E. Scott, T. Labotka, and C. Mora for useful discussions. Finally, we gratefully acknowledge R. Clayton for supplying the isotopic data used in this study, and the various institutions listed in Table 1 for supplying CM samples for analysis. (?) This paper is the Hawaii Institute of Geophysics and Planetology publication no. ??, and the School of Ocean and Earth Science and Technology Publication no. ?? (?).

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APPENDIX

Determining Mineralogic Alteration Indices

The following listing is presented to facilitate calculation of mineralogic alteration indices:

- 1) Thin sections of CM chondritic meteorites are selected for analysis.
 - a) Avoid terrestrially weathered samples, such as those exhibiting pervasive rust.
 - b) To lessen the possibility of analyzing non-phyllsilicate phases that may occur in the third dimension, thin sections are ideally ground to thicknesses of less than ~25 μm . If custom grinding is not feasible, look for sections that thin significantly toward the sample edge.
 - c) When possible, multiple thin sections of an individual CM chondrite should be obtained for analysis.
- 2) Electron microprobe analyses of CM matrix material are collected.
 - a) Survey the matrix material by electron backscattered imaging to characterize the grain size distribution, textures, and mineral associations of the sample.
 - b) Select a small region (~5 μm across) of the matrix that appears to consist largely of phyllosilicates. Collect an EDS spectra of this area to demonstrate a minimal chemical contribution by non-phyllsilicate grains. Although some S will appear in almost every analysis, those spectrum with minimal peaks for S, Ca, and Cr generally produce the best analytical results for the purpose of calculating mineralogic indices.
 - c) Standard microprobe techniques are employed to collect a chemical analysis of the matrix material selected in 2b. See analytical techniques section above for recommended microprobe operating conditions.

d) Alternate between selecting matrix points (2b) and evaluating their chemical composition (2c) until a representative selection of matrix material (2a) has been analyzed. Try to collect at least 40 points, since many of these analyses will probably be eliminated by screens.

3) Several initial screens are employed to ensure the quality of individual analyses.

a) Although microprobe analyses of CM matrices typically yield low totals, any analysis that does not produce a total of at least 83 wt. % oxides should be excluded from further consideration.

b) Analyses containing more than 0.5 wt. % Ca, Cr, or Na should be discarded due to an increased probability that carbonates, oxides, or non-serpentine phyllosilicates have been included in the analysis. Likewise, no more than 2 wt. % S is acceptable.

4) After reducing the data, values of mineralogic alteration index are calculated from atomic proportions according to the following procedure.

a) Idealized sulfide grains are subtracted from each analysis. For simplicity, all S is assumed to be in the form of $(\text{Fe,Ni})_{0.877}\text{S}$. This is consistent with previous assertions that Ni-bearing pyrrhotite is the predominant sulfide phase in CM matrices (ZOLENSKY et al., 1993). We define the number of Fe and Ni atoms that combine with all of the available S according to the following relationships :

$$\text{Fe} = (0.875 * \text{S}) - \text{Ni}$$

$$\text{Ni} = 0.1\text{S}.$$

The number of Fe, Ni, and S atoms that are incorporated into the idealized pyrrhotite are then subtracted from the original atomic proportions for each of these elements.

b) Sulfide-corrected cation totals are recalculated to 5. This is accomplished by multiplying the atomic proportion of each element by a correction factor, $5/(\text{sulfide-corrected cation total})$. This step involves the fundamental assertion that the sulfide-corrected atomic proportions reflect a stoichiometric serpentine composition.

c) New screens are imposed on the recalculated cation values to eliminate analyses that do not produce reasonable serpentine compositions. Since serpentine contains a maximum of 2 Si atoms, for example, all analyses with greater than 2 atoms of Si are excluded from further consideration. In addition, the proportions of all analyzed cations are used to determine if the resultant serpentine composition is reasonably charge-balanced. Although serpentine are seldom perfectly stoichiometric, we require that all analyses produce a total cation charge that ranges between ~13.5 and 14.

d) The number of Fe^{3+} and Fe^{2+} atoms in each analysis are estimated by demanding that the general stoichiometric requirements of serpentine, $(\text{Fe}^{2+}, \text{Mg})_{3-x}(\text{Al}, \text{Fe}^{3+})_x(\text{Si}_{2-x}(\text{Al}, \text{Fe}^{3+})_x\text{O}_5(\text{OH})_4)$, are fulfilled. By assuming that a loss of Si is accommodated by coupled substitutions that exhaust all of the available Al, the requisite number of Fe^{3+} atoms is approximated by:

$$\text{Fe}^{3+} = (2 * (2 - \text{Si})) - \text{Al}.$$

Calculated Fe^{3+} values are then subtracted from the total number of Fe atoms to give the number of Fe^{2+} atoms that remain.

e) The mineralogic alteration index is defined as the average value of $2 - (\text{Fe}^{3+}/(2\text{-Si}))$ for the matrix analyses in an individual CM chondrite. Ratios of $\text{Fe}^{3+}/(2\text{-Si})$ for individual analyses should range between 0 (Mg-serpentine) and 2 (cronstedtite). Any individual ratios that do not fall within this range are excluded from the calculation of the average $\text{Fe}^{3+}/(2\text{-Si})$ value for the sample. Ideally, at least 20 matrix analyses should be averaged into the mineralogic alteration index, but this is not always possible.

FIGURE CAPTIONS

Figure 1: An olivine grain in the matrix of Cold Bokkeveld is partially altered to serpentine phases. Irregularities in the olivine surface probably resulted from the penetration of alteration fluids along the original fracture planes of the host olivine.

Figure 2: Comparison between proposed CM alteration parameters. Plots of the mineralogic alteration index number versus a) the modal proportion of isolated matrix silicates, and b) the volume percent of chondrule alteration illustrate the continual production of phyllosilicates as anhydrous silicate phases are systematically consumed during progressive CM alteration. Meteorite abbreviations are given in Table 1.

Figure 3: The effects of progressive CM alteration on bulk chemical and isotopic compositions are evaluated. a) The H content of individual CM chondrites increases with progressive CM alteration as more hydrous phyllosilicates are produced. b) A negative correlation between the bulk ^{36}Ar content and the alteration index number indicates a significant loss of trapped planetary gases with increasing CM alteration. c) A plot of $\delta^{18}\text{O}$ vs. $\delta^{17}\text{O}$ for bulk CM samples provides another check on the order of increasing CM alteration predicted by our model because $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ values increase with advancing phyllosilicate production at low temperatures (CLAYTON, 1993). In accordance with our model predictions, the oxygen isotopic data indicate that Murchison (MC) and Bells (BL) experienced minimal alteration, while Cold Bokkeveld (CB) was extensively altered. Data for the other CM samples are poorly resolved. Locations of the terrestrial mass fractionation (TF) and CAI mixing lines (CAI) are shown for

comparison. Meteorite abbreviations are given in Figure 2. All isotopic data were graciously supplied by R. Clayton.

Table 1. CM Chondrites Studied.

Samples	Abbreviations	Thin Section I.D.s	Source
Bells	BL	B3; A3	Amer. Museum Natural History
Boreskino	BO	6495-1	Smithsonian Institute
Cochabamba	CC	1; 2	Nat. History Museum, Vienna
Cold Bokkeveld	CB	1727a	Nat. History Museum, London
Mighei	MI	3; 4; 5	Field Museum
Murchison	MC	E; H	Field Museum
Murray	MY	2; 3	Field Museum
Nogoya	NG	1; 2	ASU Center for Meteoritics
Pollen	PO	3	Nat. History Museum, London

Table 2. Serpentine Formulae*

	Chrysotile	
Mg-Bearing Members	Lizardite	$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$
	Antigorite	
	Cronstedtite	$\text{Fe}^{2+}_2\text{Fe}^{+3}(\text{SiFe}^{+3})\text{O}_5(\text{OH})_4$
Fe-Bearing Members	Greenalite	$(\text{Fe}^{+2}, \text{Fe}^{+3})_{2-3}\text{Si}_2\text{O}_5(\text{OH})_4$
	Ferroan	
	Antigorite	$(\text{Mg}, \text{Fe}, \text{Mn})_3(\text{Si}, \text{Al})_2\text{O}_5(\text{OH})_4$
Intermediate Members	Amesite	$\text{Mg}_2\text{Al}(\text{SiAl})\text{O}_5(\text{OH})_4$
	Berthierine	$(\text{Fe}^{2+}, \text{Fe}^{3+}, \text{Mg})_{2-3}(\text{Si}, \text{Al})_2\text{O}_5(\text{OH})_4$

* after ZOLENSKY and MCSWEEN (1988).

Table 3. Estimated Chemical Formulae of Average CM Matrix Serpenines, and Mineralogical Alteration Indices (M.A.I.)

Samples	Si	Al	Cr	Fe(2+)	Fe(3+)	Mn	Mg	Ca	Na	Ni	M.I.
BL (25)	1.62 (.10)	0.17 (.03)	0.02 (0)	0.97 (.26)	0.59 (.20)	0.01 (0)	1.50 (.24)	0.04 (.05)	0.02 (.02)	0.04 (.03)	1.54 (.14)
BO (14)	1.73 (.13)	0.17 (.03)	0.01 (.01)	0.91 (.18)	0.36 (.25)	0.01 (0)	1.74 (.28)	0.02 (.02)	0.02 (.01)	0.02 (.02)	1.27 (.29)
CB (24)	1.77 (.13)	0.20 (.10)	0.01 (.01)	0.9 (.27)	0.25 (.22)	0.01 (0)	1.79 (.33)	0.01 (.01)	0.02 (.02)	0.02 (.02)	0.97 (.45)
CC (14)	1.58 (.12)	0.27 (.06)	0.02 (.01)	1.07 (.18)	0.58 (.19)	0.01 (0)	1.34 (.27)	0.02 (.01)	0.03 (.01)	0.08 (.02)	1.35 (.13)
MC (14)	1.58 (.19)	0.15 (.03)	0.01 (0)	0.91 (.29)	0.69 (.37)	0.01 (0)	1.59 (.40)	0.01 (.01)	0.02 (.03)	0.03 (.02)	1.57 (.20)
MI (19)	1.64 (.14)	0.25 (.10)	0.02 (.02)	0.83 (.21)	0.47 (.21)	0.01 (.01)	1.71 (.27)	0.01 (.01)	0.03 (.02)	0.02 (.03)	1.23 (.34)
MY (11)	1.56 (.14)	0.24 (.11)	0.01 (.01)	1.17 (.43)	0.65 (.24)	0.01 (0)	1.17 (.41)	0.08 (.14)	0.05 (.02)	0.04 (.03)	1.43 (.23)
NG (14)	1.80 (.10)	0.17 (.06)	0.02 (.02)	0.58 (.18)	0.23 (.19)	0.01 (0)	2.12 (.23)	0.01 (0)	0.03 (.01)	0.02 (.03)	1.03 (.34)
PO (10)	1.65 (.07)	0.18 (.02)	0.01 (0)	1.16 (.13)	0.52 (.13)	0.01 (0)	1.38 (.19)	0.01 (0)	0.03 (.01)	0.03 (.01)	1.47 (.13)

Atomic formulae were derived from microprobe analyses that have been corrected for small amounts of sulfide, using the procedure described in the Appendix. The number in parentheses beside the sample abbreviation refers to the number of analyses averaged. Other numbers in parentheses are 2 σ standard deviations, which illustrate the range of matrix serpentine compositions in individual CM chondrites.



Fig. 1

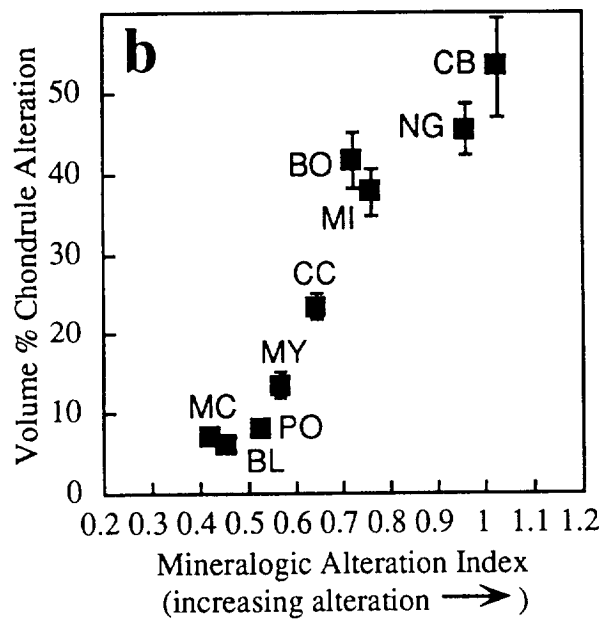
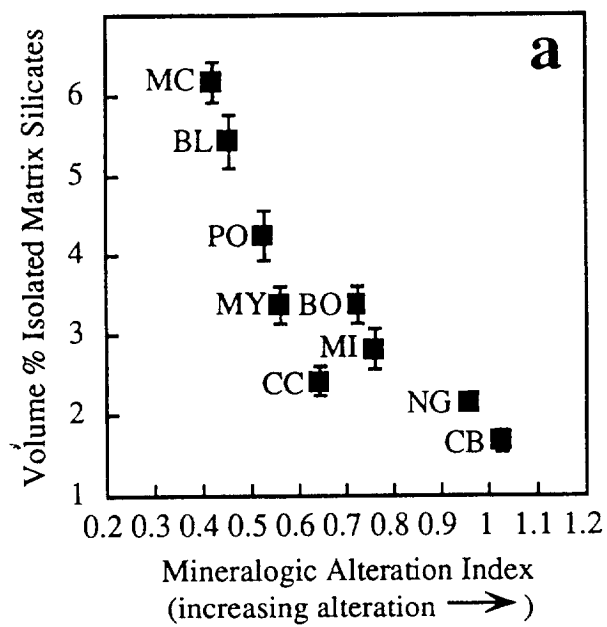


Fig 2

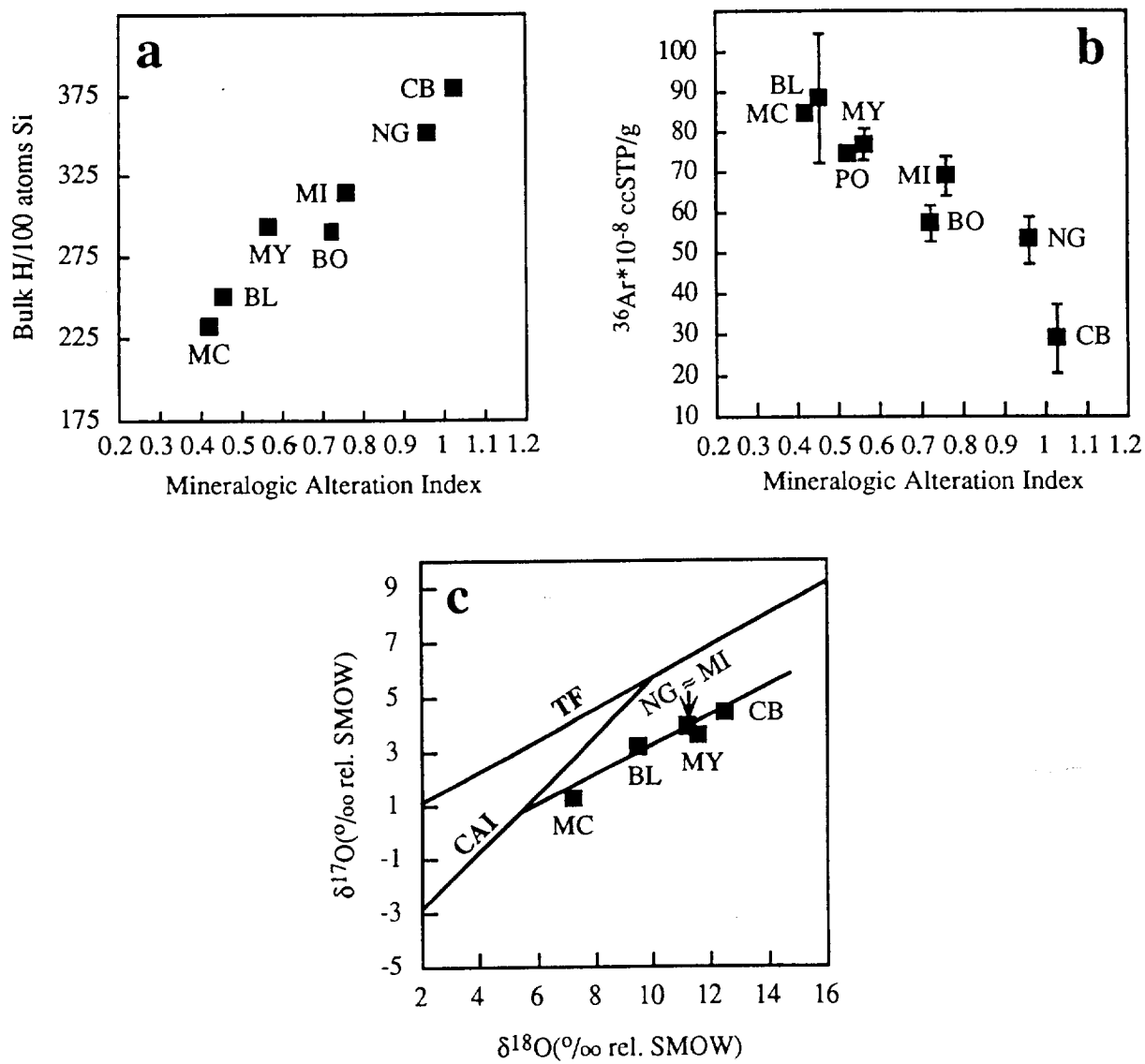
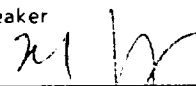
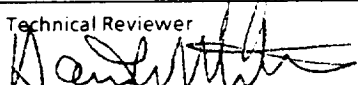

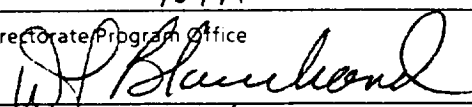

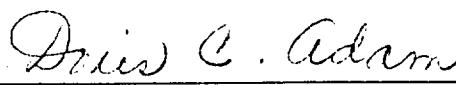
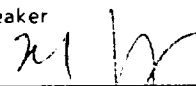
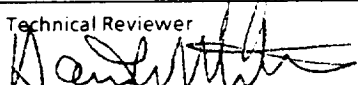

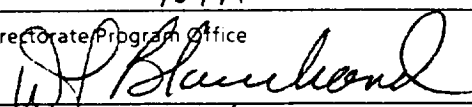

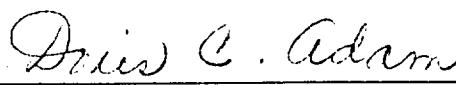
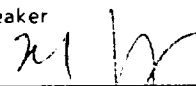
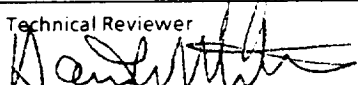

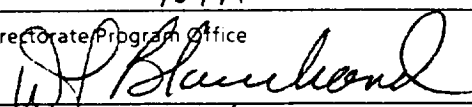

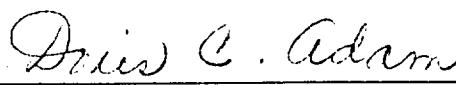


Fig 3

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